

Application No.: 09/810,743

Case No.: 51932US017

Remarks

Reconsideration and continued prosecution of the present application is respectfully requested. No amendments to the claims are being presented in this Response. Claims 1-8 remain pending.

§ 103 Rejections

Reconsideration of the Examiner's rejection of claims 1-8 under 35 U.S.C. § 103(a) as being unpatentable over U.S. 5,808,798 (Weber et al.) in view of U.S. 4,348,349 (Kurtz) is respectfully requested.

In order to support a prima facie case of obviousness, a proposed combination of references must teach or suggest each and every element of the claimed invention. In the present case, however, the proposed combination of Weber et al. and Kurtz does not teach or suggest a method of the type claimed wherein the coPEN and PMMA resins are coextruded at a melt temperature of about 260°C (independent claim 1) or at a melt temperature not below about 250°C (independent claim 4). This combination of references also does not teach the limitation in claim 1 (and in dependent claim 5) that "the birefringence of the coPEN resin is reduced by about 0.02 units or less compared to the birefringence of a homopolymer PEN resin for a given draw ratio."

The Examiner concedes that the proposed combination of references does not teach the claimed extrusion melt temperature or the recited birefringence of the coPEN resin. However, with respect to the extrusion melt temperature, the Examiner argues variously (1) that this aspect would have been within the skill level of the art, (2) that the extrusion temperature is a result effective variable that would have been readily determined through routine experimentation, and (3) that this factor would have been readily chosen based on the degradation temperature of the resin. With respect to the recited birefringence of the coPEN resin, the Examiner argues (4) that this aspect would have been within the skill level of the art, (5) that the extrusion temperature is a result effective variable that would have been readily determined through routine experimentation, and (6) that "it is well known in the art that the birefringence of a PEN resin is reduced to some extent by copolymerizing the homopolymer to make coPEN".

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With respect to points (1), (2), (4) and (5) (which relate to routine experimentation and the level of skill in the art), Applicants respectfully note that the Examiner is applying an incorrect legal standard in assessing the patentability of the present invention. The Examiner is respectfully reminded that 35 U.S.C. § 103(a), under which the present rejection is being made, specifically dictates that “[p]atentability shall not be negated by the manner in which the invention was made.” Hence, it does not matter for the purposes of assessing patentability whether a claimed invention was, or could have been, arrived at through routine experimentation, or whether such experimentation was or would be “within the skill level of the art”. Rather, the relevant inquiry is whether *the claimed subject matter as a whole* is taught or suggested by the prior art.

M.P.E.P. § 706.02(j) describes the criteria to be used in determining whether the claimed subject matter as a whole is obvious:

To establish a *prima facie* case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. Second, there must be a reasonable expectation of success. Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations. The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be found in the prior art and not based on applicant's disclosure. ... The initial burden is on the examiner to provide some suggestion of the desirability of doing what the inventor has done.

In the present case, the cited prior art does not teach or suggest the claimed extrusion melt temperatures. Moreover, the Office Action fails to explain why it would be desirable to modify the methodology of the cited art through the use of higher melt extrusion temperatures so as to arrive at the presently claimed methods (indeed, as explained below, such extrusion melt temperatures would not be considered desirable, absent the teachings of the present application, because the PMMA would be expected to degrade significantly under these conditions; for this reason, there would also be no reasonable expectation of success in making the proposed modification). Hence, the Office Action fails to establish a *prima facie* case of obviousness.

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With respect to point (3), Applicants respectfully note that consideration of the degradation temperature of the resin would not have led one skilled in the art to utilize an extrusion melt temperature within the ranges recited in the claims. To the contrary, it is well known in the art that PMMA quickly degrades at temperatures at or above 250°C. It is largely for this reason that, as noted by Applicants at page 21, lines 25-26 of the present (substitute) specification, “polymethyl methacrylate (PMMA) is typically extruded at a temperature below about 250°C”.

This fact is underscored by P. Nising et al., “On The Degradation and Stabilization of Poly(Methyl Methacrylate) in a Continuous Process”, Chem. Eng. Technol. 26, 599-604 (2003) 5, a copy of which is attached hereto as EXHIBIT A. As shown in FIGS. 1 and 5 of this reference, PMMA undergoes degradation of about 30% relative mass loss at 250°C, due to the presence in the polymer of head-to-head bonds and unsaturated end groups (zones I and II in the graph, respectively). The amount of degradation goes up from there as temperature rises. Thus, at 260°C (claims 1 and 8), PMMA undergoes degradation of about 40% relative mass loss, while at 275°C (claim 7), PMMA undergoes degradation of about 45% relative mass loss.

In light of the above, one of ordinary skill would not have found it obvious to coextrude coPEN and PMMA at the claimed temperature ranges, because such person would have expected a substantial amount of degradation to occur in the PMMA at these temperatures. Indeed, as noted at the paragraph beginning at page 21, line 22 of the present specification, it was Applicants’ surprising discovery that PMMA can be coextruded with PEN using melt temperatures as high as 275°C, provided that certain precautions are taken (e.g., that design considerations are made in the PMMA melt train to minimize the potential for stagnation points in the flow, and to hold to a minimum the overall residence time in the melt of the PMMA).

With respect to point (6), Applicants respectfully submit that the Office Action is misconstruing the claim limitation at issue. In particular, claims 1 and 5, which contain the relevant limitation, do not require a reduction in the birefringence of the coPEN. Rather, these claims require that “the birefringence of the coPEN resin is reduced by about 0.02 units *or less compared to the birefringence of a homopolymer PEN resin* for a given draw ratio”. Hence, these claims require, in essence, that, at a given draw ratio, the birefringence of the coPEN is

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not reduced substantially relative to the birefringence of PEN. The Examiner's own remarks underscore the unobviousness of this result (i.e., that "it is well known in the art that the birefringence of a PEN resin is reduced to some extent by copolymerizing the homopolymers to make coPEN").

Further support for Applicants' contention that this feature of the claimed invention is unobvious can be found in U.S. Patent Application 2003/0164914 (Weber et al.) (a continuation of a number of issued U.S. Patents, including US 6,543,153 and 6,025,897), which is commonly assigned and relates to a similar technology. As noted in EXAMPLE 1 of that application, single layer films of PEN and coPEN were examined in both a stretched and unstretched state. The birefringence of the PEN film was observed to increase 0.23 at a 5:1 stretch ratio compared to the unstretched film, while there was no observable difference in the birefringence of the coPEN film in its stretched or unstretched states. This reference thus demonstrates that coPEN formulations are known to the art wherein the birefringence of the coPEN is dramatically different from the birefringence of PEN. Since the Examiner has not established that coPEN formulations are known to the art wherein the birefringence of the coPEN is similar to the birefringence of PEN (and in particular, wherein "the birefringence of the coPEN resin is reduced by about 0.02 units *or less compared to the birefringence of a homopolymer PEN resin* for a given draw ratio"), the Office Action has failed to establish a prima facie case of obviousness of the pending claims.

With respect to the step of casting the multilayer web onto a casting roll to form a cast multilayer film, the Examiner apparently relies upon Kurtz for this teaching. Kurtz, however, does not remedy the deficiencies in the primary reference (Weber et al.) pointed out above. The rejection of pending claims 1-8 should therefore be withdrawn.

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Conclusion

Applicants submit that the pending claims 1-8 are allowable over the art of record for the reasons given.

No fee is believed to be due by submission of this paper. However, if this belief is in error, please charge any required fee to Deposit Account No. 13-3723.

Respectfully submitted,

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EXHIBIT A:
On The Degradation and Stabilization of Poly(Methyl Methacrylate) in a Continuous Process”,
Chem. Eng. Technol. 26, 599-604 (2003) 5

On the Degradation and Stabilization of Poly(Methyl Methacrylate) in a Continuous Process

By Philip Nising, Thomas Zeilmann, and Thierry Meyer*

Thermally unstable polymers such as poly(methyl methacrylate) are degraded considerably during industrial processing. This degradation and its reduction to a minimum have been investigated in both lab and continuous pilot-scale experiments. A three-step degradation mechanism, starting at 180 °C, was proved by Thermogravimetical Analysis (TGA) and a kinetic approach to describe it was derived. The knowledge of this degradation behavior was then applied to a pilot-scale process with a production rate of 10 kg/h and the process yield loss during the devolatilization step was investigated. Using heat stabilizers, the overall process yield could be improved by 10 %, whereas the residual organic volatiles concentration (VOC) was drastically reduced to values below 1000 ppm. In order to preserve the molecular weight of the final product these stabilizers were added into the process, separately, at the end of the polymerization reaction but before the devolatilization step.

1 Introduction

With the increasing demand for lower molecular weight poly(methyl methacrylate) (PMMA) for extrusion and injection molding, continuous polymerization processes become more and more important for its production. Solution polymerization processes are required in order to ensure a pumpable mass flow in the plant. At the end of the process, solvents and residual monomer must be removed from the polymer melt in a devolatilization step. This is usually realized by heating up the solution followed by a strong pressure drop (flash), thus vaporizing the volatile fraction. Until now, since PMMA has a rather poor thermal stability, this last process step limits the overall yield significantly by degrading polymer chains back, mainly, into monomer.

For sufficient devolatilization, the minimum preheating temperature, depending on the reactor setup, was determined by pilot-scale experiments to be 280 °C. At this temperature, a yield loss of 10 % is observed that can no longer be neglected in continuous production. This yield loss is temperature dependent, with 4 % at 240 °C and going up to 12 % at 300 °C. It is therefore necessary to first investigate the degradation behavior of unstabilized PMMA at temperatures similar to the one in the production process as well as to find strategies for stabilizing the polymer without changing its product properties.

2 Non-oxidative Degradation of PMMA

Poly(methyl methacrylate) degrades back into monomer above 180 °C in three different steps by an unzipping reaction. The monomer yield for PMMA can be up to 90 % [1]. A typical degradation curve measured by TGA is presented in

Fig. 1, where the different degradation steps are clearly visible.

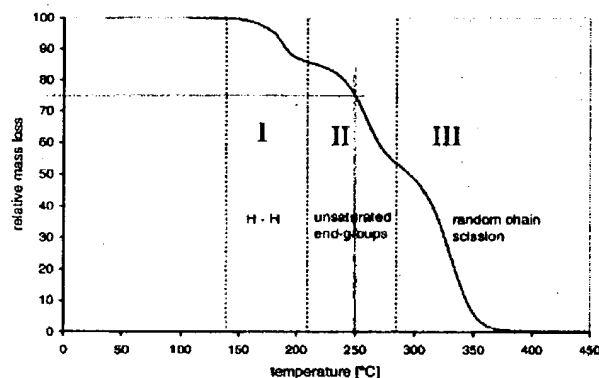


Figure 1. TGA curve (0.5°C/min) for non-commercial PMMA from a batch experiment, radically polymerized at 130 °C with 30 mmol/L DBPO.

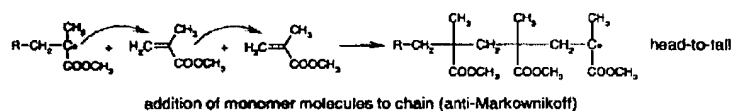
As already shown by [2] and verified by our own measurements, these three steps are due to three different types of bonds within the polymer chains, formed by different reactions during radical polymerization, Fig. 2:

- Regular polymer backbone bonds (r-c), zone III
- Unsaturated end groups (d-b), zone II
- Head-to-Head-bonds (h-h), zone I

The polymer chain propagation consists of the addition of one monomer unit to the growing macroradical, in general by a "head-to-tail" mechanism. This means that, stereochemically, the monomer is always bound to the polymer chain by its least substituted carbon. Thus, the relatively big -COOMe group is always ahead of the growing chain. This "head-to-tail" propagation leads to a uniform backbone chain with C-C bonds that are thermally stable up to 300 °C. Above this, they break up statistically, known as random chain scission. The unsaturated end groups and the head-to-head bonds are formed mainly by termination reactions. Apart from termination by other radical species, two active polymer chains can also terminate themselves in two different ways: either by disproportionation, leading to a double bond in one chains

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chain propagation



termination reactions

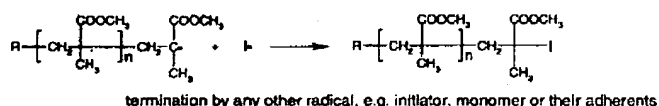
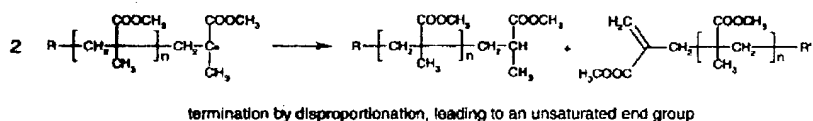
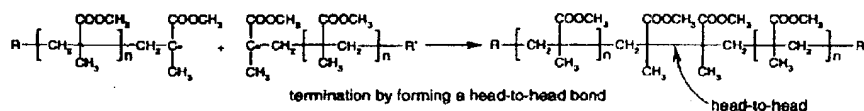


Figure 2. Important reaction steps of MMA polymerization.

end group (thermally stable up to 250 °C), or by head-to-head coupling. In the latter case, a thermally very unstable C-C bond is formed, which breaks easily at temperatures as low as 180 °C. Depending on the thermal bond stability, the polymer chain is activated by bond scission at the corresponding temperature and is completely depolymerized by an unzipping reaction.

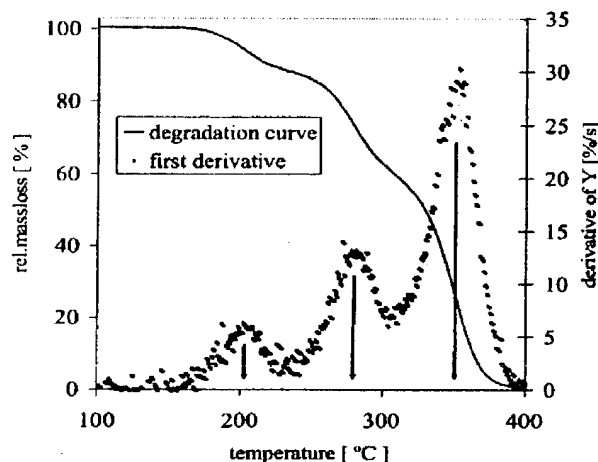
For further kinetic investigation of the degradation reaction, it is assumed that the "activation reactions" as described above are the rate determining steps, whereas the unzipping is so fast that it can be neglected in the calculation. However, the kinetic parameters E_A (apparent activation energy) and k_0 (pre-exponential factor) are needed. They are obtained by dynamic thermogravimetric measurements in combination with a calculation scheme [3]: The thermogravimetric analysis is carried out for samples ($m \approx 20$ mg) of the same polymer with 10 different heating rates (0.5 °C/min, 1.0 °C/min, 1.5 °C/min, 2.5 °C/min, 3.0 °C/min, 4.0 °C/min, 5.0 °C/min, 7.5 °C/min, 9.0 °C/min, and 10.0 °C/min; at higher heating rates, the three steps become blurred and a clear division into three parts impossible). Then, the first derivative of each degradation curve (specific mass loss against temperature) is calculated (shown for one heating rate in Fig. 3, as example).

The kinetic values can be obtained using Eq. (1) by determining T_m (temperatures at each curve's extreme values).

$$\ln \left[\frac{\partial T / \partial t}{T_m^2} \right] = \ln \left[k_0 \frac{R}{E_A} \right] - \frac{E_A}{R} \cdot \frac{1}{T_m} \quad (1)$$

The graphical resolution of Eq. 1 is presented in Fig. 4 and the numerical values can be found in Tab. 1, for all the

different heating rates, where it appears that the apparent activation energy for the h-h and d-b bonds are equal, but slightly smaller than for the r-c scission.

Figure 3. Degradation curve and its first derivative with T_m 's for maximum weight loss.Table 1. Kinetic values for the three different degradation initiation reactions, relative errors and coefficient of determination, R^2 .

Bond type	E_A [kJ/mol]	\pm [%]	k_0 [s ⁻¹]	\pm [%]	R^2
h-h	$-2.08 \cdot 10^2$	10	$3.4 \cdot 10^{19}$	12	0.9495
d-b	$-2.07 \cdot 10^2$	4	$9.3 \cdot 10^{15}$	5	0.9886
r-c	$-2.18 \cdot 10^2$	5	$2.4 \cdot 10^{14}$	7	0.9775

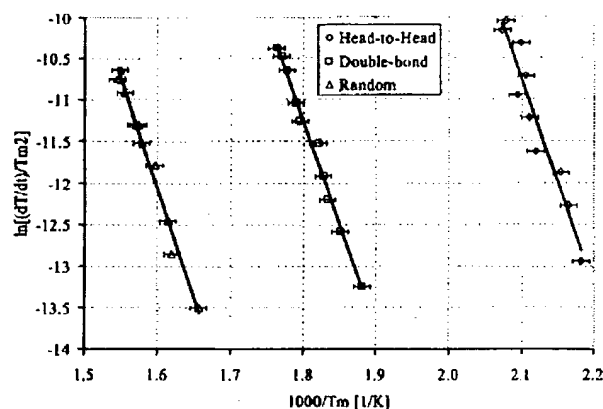


Figure 4. Quasi-Arrhenius plot for the three degradation steps of PMMA.

Using these kinetic values, PMMA degradation can be simulated for the three steps using a basic first order kinetic approach.

$$\frac{dm}{dt} = -km \rightarrow \frac{dm}{m} = -k dt \quad (2)$$

In order to gain a direct correlation between mass and temperature, the term dt has to be substituted by:

$$dt = \frac{1}{dT/dT} dT \quad (3)$$

where dT/dt represents the heating rate. By including the formal kinetics for k , the following expression is derived:

$$\frac{dm}{m} = -k_0 \exp\left(-\frac{E_A}{RT}\right) \frac{1}{dT/dt} dT \quad (4)$$

leading, after integration, to the following equation:

$$\ln \frac{m}{m_0} = -\int k_0 \exp\left(-\frac{E_A}{RT}\right) \frac{1}{dT/dt} dT \quad (5)$$

The integral on the right side can be solved explicitly by substitution. For our purpose, however, an approximation by summation is sufficient (for a more detailed description see [4]):

$$\frac{m}{m_0} = \exp\left(-\sum_T k_0 e^{-\frac{E_A}{RT}} \frac{1}{dT/dt} \Delta T\right) \quad (6)$$

This equation has to be applied separately to each degradation step by using the corresponding kinetic values from Tab. 1. However, the values for m_0 are still missing. These constitute the mass of polymer that is degraded during the respective step. This amount could be taken from a preceding TGA experiment or estimated from comparable polymer samples. The association between the simulated curve and the calculated reaction rates (Figs. 5 and 6) indicates that the assumed reaction mechanism correctly describes the experiments.

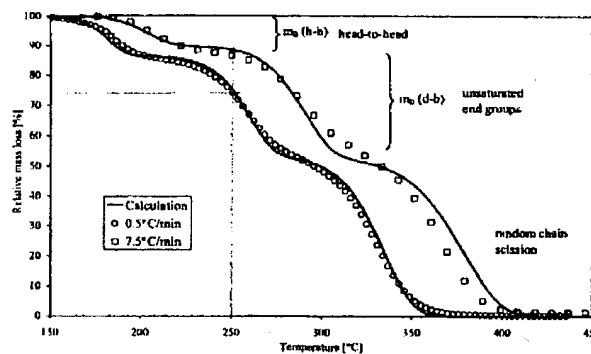


Figure 5. Calculated degradation curve compared to experimental data.

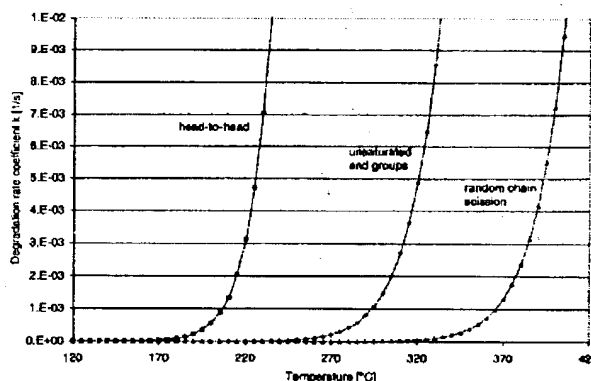


Figure 6. Degradation rate constants, k , evolution with temperature.

The degradation rate-constants are highly temperature dependent and do not overlap each other, as presented in Fig. 6. In Fig. 5, it can clearly be seen that for higher heating rates, the three degradation steps become blurred, and thus, that the calculated curve deviates from the experimental curve in the interfaces between each step since it does not take the inertia of the measurement into account.

3 Thermal Stabilization in a Continuous Pilot-Scale Process

Efficient devolatilization in continuous polymerization processes requires temperatures above 240 °C in the polymer melt. As seen above, at these temperatures the process productivity is considerably diminished by degradation. In order to improve the overall yield and meet product specifications, the polymer must be stabilized for elevated temperatures. The stabilization is usually realized by adding up to 1 % (w/w) of a thiol chain transfer agent to the monomer, which influences the termination behavior during the polymerization reaction and at the same time the molecular weight distribution. However, the necessary thiol concentration, in order to avoid degradation at temperatures below 200 °C, was found to be so high (>3 % (w/w)) that the final products

molecular weight fell to intolerably low values (at least one order of magnitude).

In order to overcome this problem, stabilizers were added after the polymerization reaction had taken place, a so to speak "stabilization after reaction". Therefore, a special setup was constructed, consisting of a high-pressure LC-pump to inject the stabilizer solution into the tubular reactor and several static mixing elements to guarantee sufficient mixing of the less viscous solution with the highly viscous polymer melt, as depicted in Fig. 7.

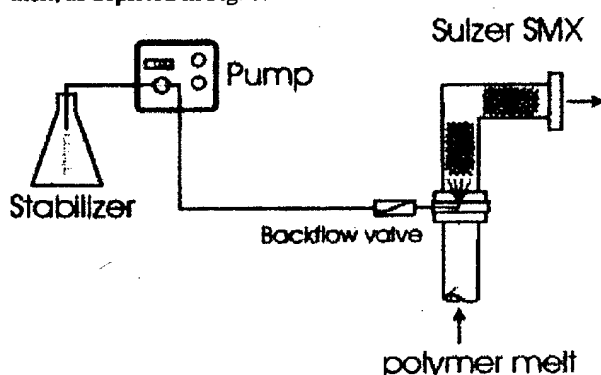


Figure 7. Post reaction injection assembly.

With the knowledge of the degradation mechanism from section 2, i.e., the fact that it is initiated by the breaking up of a

chain bond into radicals, different kinds of stabilizers were chosen for further investigation. It was assumed that radical scavengers would act as stabilizers as they catch not only free radicals in the polymer melt, which can attack intact chains, but they were also supposed to bind chain radicals already in their statu nascendi, i.e., before they unzip. Yet, due to the wide product spectrum, it was necessary to restrict oneself to certain promising products. First of all, *n*-dodecanethiol, a thiol commonly used as chain transfer agent, was chosen as reference since its ability to transfer a proton to another radical can also work for the prevention of unzipping. Secondly, a newly developed stabilizer from Ciba Specialty Chemicals Inc. was used. It is based on an aromatic lactone and is also able to transfer a proton, itself becoming a highly resonance stabilized radical. Thirdly, for comparison, conventional organic phosphites were tested for their ability as stabilizers. An overview of the different stabilizers is provided in Tab. 2.

A scheme of the pilot plant is depicted in Fig. 8.

It generally consists of three units: one for the preparation of the feed solutions (A), the reactor unit with a combined loop and tubular reactor (B), and the devolatilization unit (C) with preheater and vacuum chamber.

The feed solution consists of the monomer (MMA), a solvent (Ethylbenzene $\approx 10\%$) and a peroxide initiator (DiTerbutylBenzoylPeroxide 15 mol m^{-3}). It is degassed before use under light vacuum and nitrogen. The throughput

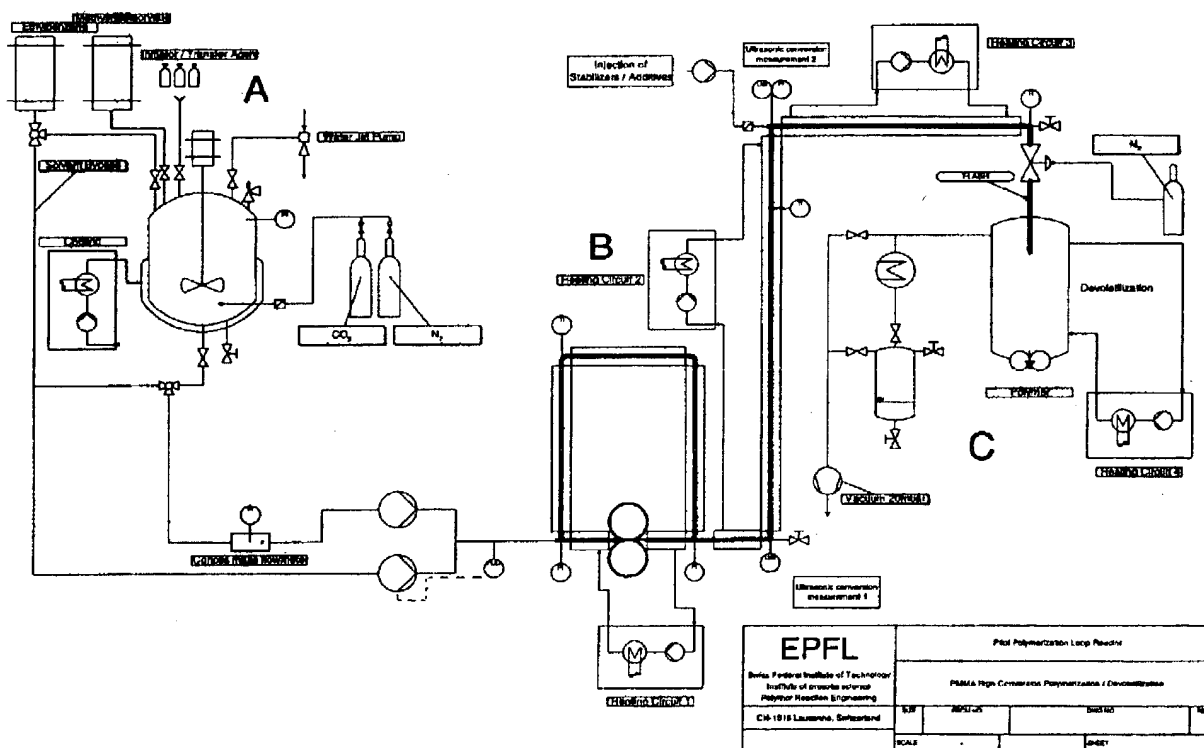


Figure 8. P&I diagram of the pilot polymerization plant.

in the reactor can be varied from 2–10 kg/h and controlled by a Coriolis mass flow meter. The reaction temperature is set to 120–140 °C and the devolatilization temperature to 240–250 °C with an absolute pressure of around 20 mbar in the devolatilization tank. The final product is then continuously discharged from the vacuum chamber with a heated polymer gear pump.

Due to current secrecy agreements with industrial partners, more details cannot be given.

Table 2. Commercial heat stabilizers used (product names are registered trademarks).

product name	molecular structure	properties
ADK STAB HP-10 (ADEKA Palmarole)		$M_w = 583$ g/mol mp = 148 °C white powder
ADK STAB PEP-36 (ADEKA Palmarole)		$M_w = 633$ g/mol mp = 238 °C white powder FCA-approved
Irgafos 38 (Ciba Specialty Chemicals Inc.)		$M_w = 514$ g/mol mp = 89–92 °C white powder FCA-approved
Irgafos 168 (Ciba Specialty Chemicals Inc.)		$M_w = 514$ g/mol mp = 89–92 °C white powder FCA-approved
Irgafos XP30 (Ciba Specialty Chemicals Inc.)		$M_w = 350$ g/mol white powder FCA-approved
n-Dodecanethiol		

4 Results

Both conversion and residual volatile concentrations were measured by Dynamic Headspace Gas chromatography with a Perkin Elmer Thermal Desorber combined with a Perkin Elmer Autosystem GC. Special attention was focused on the molecular weight distributions, which were determined by Size Exclusion Chromatography on a Viscotek TDA 300 Triple Detection SEC.

The results from stabilization after reaction were very positive. Compared with the non-stabilized samples, the overall process yield could be improved by nearly 10 % depending on the stabilizer used, Fig. 9.

Taking the throughput of common PMMA production plants into account, this constitutes a considerable increase in process efficiency. Also, as can be seen later for the residual

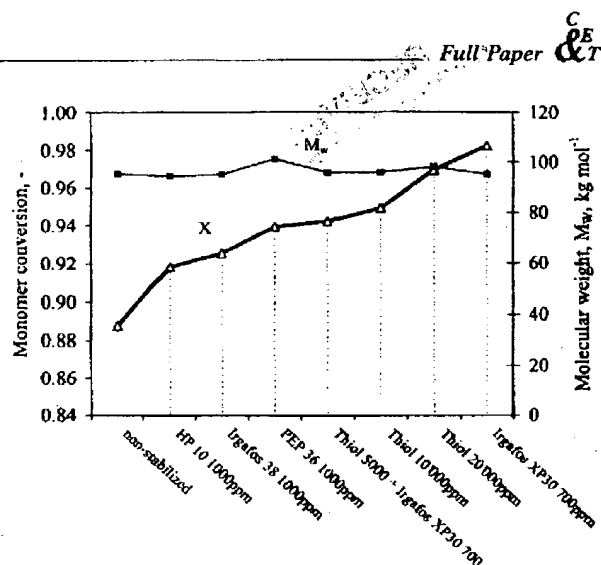


Figure 9. Increase in overall process yield by stabilization after reaction.

volatiles, there is a major difference between the basic types of stabilizers. The conventional phosphite based stabilizers only have minor effects on the thermal stability, which underlines that the main reason for thermal degradation of PMMA is not peroxides in the polymer melt. On the other hand, with the thiol and the lactone-based stabilizer, the results were remarkably better. Therefore, it can be concluded that the termination of chain radicals, during their formation, by highly reactive proton donors is the most effective strategy to stabilize PMMA in high temperature production steps.

The same results were obtained for the residual VOC. Fig. 10 shows the residual monomer and solvent concentrations for different stabilized and non-stabilized polymer samples.

By adding only 1 % thiol and between 700 and 2100 ppm of lactone stabilizer, the monomer concentration in the final polymer could be reduced by one order of magnitude, to less

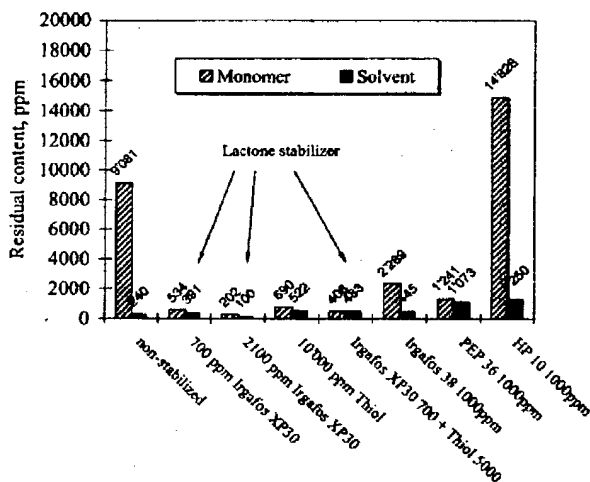


Figure 10. Residual volatiles for stabilized and non-stabilized polymer.

than 100 ppm. Again, the phosphite species were not convincing, especially as one of them (HP 10) had a deteriorating effect on the residual monomer content. The fact that the residual volatiles are so low also indicates that the stabilizers remain active after devolatilization. The reason is that even with a highly efficient devolatilization, the polymer is still exposed to high temperatures during discharge from the reactor by heated gear pumps. The monomer formed in this last step will remain in the polymer due to the immense diffusion limitation in the viscous melt. Thus, in order to achieve lowest residual volatiles concentrations, stabilization throughout the whole last "production step" (Czone in Fig. 8) is inevitable.

For all experiments with stabilizers, the molecular weight of the final product was monitored by GPC and compared to the molecular weight of the non-stabilized samples. As expected for the post-reaction stabilization strategy, there was no visible influence of the stabilizing agents on the molecular weight distribution, Fig. 9. The effect of the stabilizer on the polymer produced before devolatilization could also be proved by dynamic TGA measurements, Fig. 11.

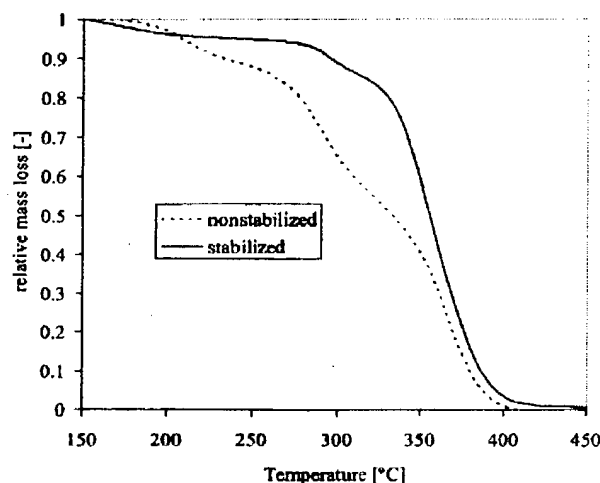


Figure 11. Non-stabilized and stabilized polymer (heating ramp 8°C/min, stabilizer Irgafos XP30, 750 ppm).

Stabilization reduces the effect of the first and second degradation but has no effect on the regular backbone scission. A non-stabilized polymer will lose approx. 50 % of its weight up to 300 °C, but a stabilized one will lose only 15 %. An appropriate stabilization improves not only the polymer quality but also economic aspects.

5 Conclusions

The TGA experiments clearly show a three-step degradation mechanism for the thermal degradation of PMMA, as already reported by other authors [2]. The kinetics for this reaction were derived and successfully confirmed. It was found that the rate-determining step is the activation of dead polymer chains at thermally weak bonds.

A new stabilization strategy for the thermal stabilization of poly (methyl methacrylate) could be established and applied to pilot plant experiments. The stabilizers are no longer added to the monomer solution feed but continuously injected into the process between the reaction and the devolatilization step. In a stabilizer screening, where several commercial stabilizers and chain regulators were tested, a special lactone based product was found to be highly efficient for PMMA stabilization.

The decrease in overall process yield by degradation in the devolatilization step could, thus, be counteracted and increased by almost 10 %. At the same time, the residual volatiles in the final product were reduced by one order of magnitude. This is an important result for product quality improvement with respect to environmental and health regulations.

Acknowledgements

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